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Generation Condition of Honeycomb Discharge
-Characteristics of the discharge of a bundle of glass capillaries-

Satoshi Sato[1], Karol Hensel[2], Hiroki Yamauchi[1],
Kazunori Takashima[1] and Akira Mizuno[1]

Abstract— **Selectivity and energy efficiency of plasma chemical processes can be improved significantly if plasma is combined with a catalyst. Generation of stable discharge plasma inside narrow capillaries of honeycomb catalyst for automobile exhaust after treatment, however, has been quite difficult. The paper presents a method to generate discharge inside honeycomb. The method uses a packed-bed discharge connected in series with honeycomb capillaries. With a DC voltage across the capillaries, streamers are extended from the packed-bed discharge into the capillaries. With this method, ionization can be made inside fine channels of honeycomb catalyst made of insulating materials. This discharge is designated as "honeycomb discharge". Electrical and optical characteristics of the honeycomb discharge generated inside a bundle of quartz capillaries are measured. Effects of the applied voltage and power, diameter of the capillaries, and composition of the gas mixture passed through the capillaries on the emission spectra of the honeycomb discharge are presented.**

***Keywords* — discharge plasma, honeycomb catalyst, honeycomb discharge**

I. INTRODUCTION

Various types of atmospheric pressure discharges have been developed for electrostatic processes and for plasma chemical processes, such as removing particles, cleaning exhaust gas and volatile organic compounds (VOCs), etc. [1-4]. In order to improve selectivity and energy efficiency of plasma chemical processes, combination of plasma and catalyst is effective. For example, catalyst pellets can be used in packed bed discharge to improve removal efficiency of nitrogen oxides and VOCs [5-6]. Honeycomb is a commonly used geometry of catalysts. It has been difficult to generate electrical discharge evenly inside a honeycomb [3]. If discharge is generated inside a honeycomb, larger surface area can be obtained with lower pressure drop for improved chemical reactions.

There are several important works recently reported. One is the superposition of surface discharge and ac discharge to obtain large discharge volume [5]. Surface discharge is generated on inner wall of a cylinder, and an ac voltage is applied between the centered electrode and the electrode placed on the inner wall for the surface discharge. The other is a sliding discharge, that has been used to cover large surface area of wings of airplane to stabilize airflow [7]. Surface discharge is generated using a pair of electrodes placed between a sheet of insulator film. The other electrode is set apart from the electrode for surface discharge on the film, and energized with negative DC. From the surface discharge, streamers are extended by the DC electric field, and cover the large surface area.

In order to ionize honeycombs consisting of fine channels, a packed-bed discharge is used in front, and DC electric field is applied across the honeycomb. This electrode configuration enables the ionization of the fine channels (1 mm square) of a honeycomb made of cordierite.

In this study, conditions for establishing plasma inside a bundle of transparent glass capillary tubes (inner diameter 1 or 2mm) that simulates the channels of honeycomb catalysts, has been studied. This is because the glow associated with the discharge can be visualized in this setup. Characteristic of a honeycomb discharge in a bundle of glass capillaries are reported.

II. INSTRUMENTATION

A. Plasma Reactor

Figure 1 illustrates the plasma reactor used in this study. A packed-bed discharge was used to form the preceding discharge. The γ-Al_2O_3 pellets (3mm) were set in a quartz glass tube (inner diameter: 26mm) with a stainless steel rod (diameter: 6mm) at the center as a discharge electrode. The

[1] Department of Ecological Engineering Toyohashi University of Technology
1-1 Tempaku-cho, Toyohashi, Aichi, 441-8580, Japan
Corresponding author, A. Mizuno: e-mail: mizuno@eco.tut.ac.jp
[2] Department of Astronomy, Earth Physics and Meteorology, Faculty of Mathematics, Physics and Informatics, Comenius University

outer surface of the quartz glass tube was covered with an aluminum sheet of 15 mm length as a ground electrode.

In this experiment, bundled thin quartz glass capillary tubes were used to simulate a honeycomb catalyst and to observe the light emission. The inner diameter of the quartz glass capillary was 1 or 2 mm. Length was 20mm. The diameter of the bundle was 26 mm and the bundle was inserted into the outer quartz glass tube. A stainless steel mesh (16mesh) was placed on the upper end of the capillaries. This stainless steel mesh was connected to a DC power supply to form the honeycomb discharge in the capillary.

In the packed-bed, discharges are generated at contacting points of the γ-Al_2O_3 pellets (3mm). The packed-bed discharge is used as the preceding discharge. When DC high voltage is applied between the bundle of quartz glass capillaries, the inside of the capillaries can be ionized, as if the AC discharge (packed bed discharge) is sliding into the capillaries by the DC electric field.

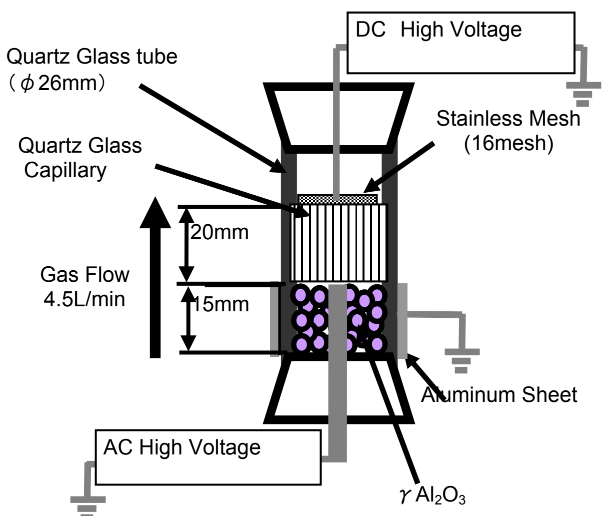


Figure 1 Schematic diagram of the reactor with bundle of capillary

B. Experimental Setup

Figure 2 illustrates the Experimental Setup used in this study. The experimental measurements were carried out at room temperature under normal pressures. An air pump placed at the downstream of this system was used to generate gas flow. The gas flow rate was 5L/min.

A DC high voltage power supply (Pulse Electronic Engineering Co., Ltd. HDV-50K3SUD) and an AC high voltage power supply (TREK Model20/20C High Voltage Amplifier) equipped with a function generator (Agilent Function/Arbitrary Waveform Generator 33220A) were used. Waveform of the applied voltage was measured using a digital oscilloscope (Tektronix TDS 2014) equipped with a high voltage probe (Tektronix P644A).

The "starting voltage of the honeycomb discharge in capillary" is defined as the voltage when the light emission of the discharge is observed inside the capillary.

The "maximum voltage" is defined as the voltage when sparking takes place and the DC high voltage cannot be applied any more.

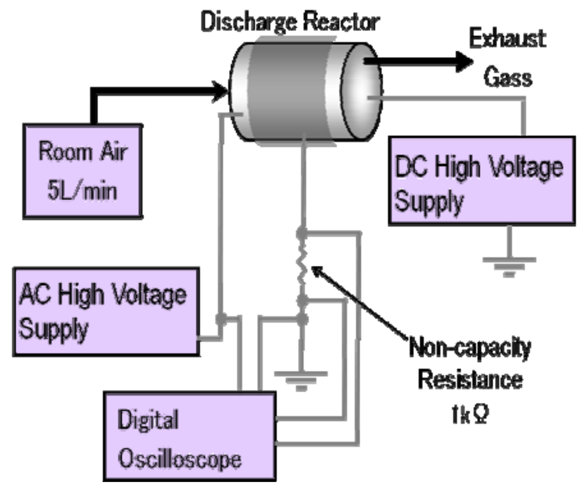


Figure 2 Experimental Setup

C. Measurement of Wave form

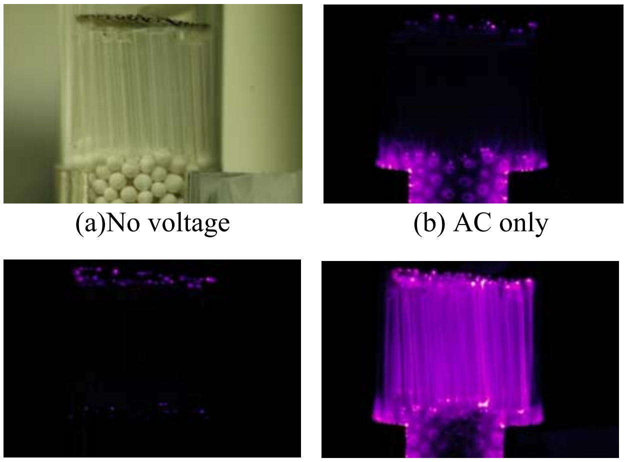
The waveform of AC high voltage was measured using a digital oscilloscope. The waveform of total discharge current was measured with a non-inductive shunt resistor. A 1k-ohm resistor was inserted in the circuit between the grounded electrode and the ground.

III. RESULT AND DISCUSSION

A. Basic Characteristics of the Honeycomb Discharge in bundle of capillaries

Figure 3 shows photographs of the discharge in the bundle of quartz glass capillaries of 1 mm inner diameter. Each photos in Figure 3 are as follows: (a): without applied voltage, (b): AC alone, (c): DC alone, (d): both AC and DC to form a honeycomb discharge in the bundle. The applied AC high voltage was $25kV_{p\text{-}p}$, 1kHz, and the DC high voltage was -11kV.

In Figure 3(b) when the AC voltage was applied, the packed bed discharge was observed. In (c) with the DC alone, light emission was observed only in the upper part of the quartz glass tube. In (d) with the AC and DC, the honeycomb discharge in the bundle of capillaries was taking place, and uniform light emission along the capillary tube was observed. Intensity of the light emission increased with increasing the DC voltage until sparking.



(a)No voltage (b) AC only

(c)DC only (d) AC and DC

Figure 3 Photographs of the honeycomb discharge in the bundle of capillary
Quartz Capillary:1mm inner diameter, 20 mm in length, AC : 25kV$_{p-p}$, 1kHz, DC: -11kV

B. The relationship between the DC and AC high voltage

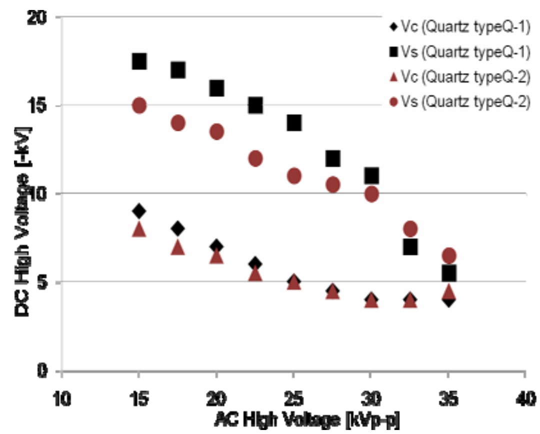
The relationship between the DC and AC high voltages to generate uniform honeycomb discharges in quartz glass capillary was measured. Figure 4 indicate the starting voltage Vc of the honeycomb discharge in the quartz glass capillary and the maximum voltage Vs (flashover voltage) of the DC voltages when the AC voltage is changed. The DC voltage is negative in Figure 4(a), and positive in (b).

The DC voltage was increased slowly from zero until sparking took place. When it was measured, the AC high voltage and the frequency were fixed. The influence of inner diameter size of the quartz glass capillary was also measured.

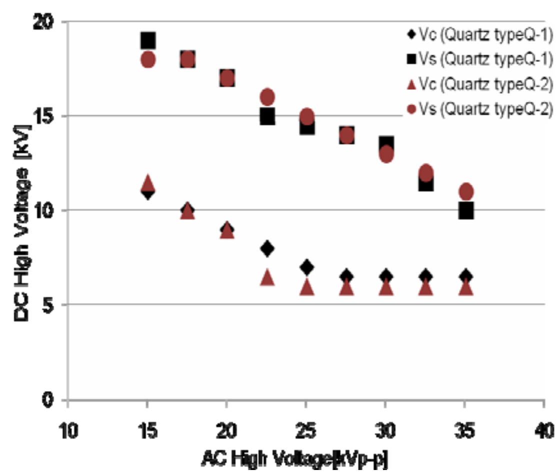
In Figures 4, Vc decreased with the increasing of AC voltage. With negative DC voltage, (Vs – Vc) was larger than that with positive DC. With negative DC, both Vs and Vc values were lower than those with positive DC.

It is well known that positive streamers tend to extend further compared to negative streamers. In this honeycomb discharge in the capillary, ionized gases in the packed-bed discharge acts as an electrode, which supplies electrons, ions, and photons more easily than metal electrodes. Therefore, more effective streamer generation and propagation could be achieved.

In the experimental condition, the honeycomb discharge took place roughly with the average DC electric field in the capillary of 3 – 8.5 kV/cm and depended on the AC voltage value. In the meantime, stronger light emission was observed from the quartz glass tube when negative high voltage was applied.



(a) Negative DC



(b) Positive DC

Figure 4 Relationship between DC and AC high voltage for generation of the uniform discharge plasma

Capillary length 20 mm, and Q-1; 1.0 mm, Q-2: 2.0 mm inner diameter
Vc: The starting voltage
Vs: The maximum voltage (the flashover voltage)

C. Waveform measurement

The waveform of AC high voltage and total current was measured.

Figure 5 shows the waveform of AC high voltage and Total current when no DC high voltage was applied. Positive and negative current were observed when the AC voltage was increasing or decreasing with time. The current pulses disappeared when the applied AC voltage reached the maximum and minimum. Because only packed-bed discharge was generated in this case, the current shown in figure 5 was typical of barrier discharges.

Figure 6 shows the waveform of the AC high voltage and the total current when the honeycomb discharge was generated by the DC application to the bundle of capillaries. When the AC voltage reached the maximum in the positive half cycle a negative current pulse was observed. This result suggests that the negative pulse current can be attributed to the honeycomb discharge.

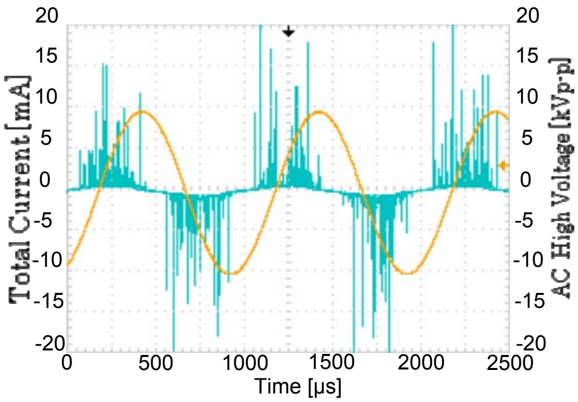


Figure 5 Waveform of AC high voltage and Total current for generation only packed bed discharge
(Negative DC high voltage 0kV, 2.0 mm inner diameter, AC high voltage 20kVp-p, 20 mm capillary)

In Figure 6, the current pulse at the peak of the ac voltage (at dV/dt = 0) was observed only in the positive half cycle of the applied voltage. This is because the capillary tubes were negatively biased and the positive voltage to the packed bed enhances the electric field along the capillary tubes.

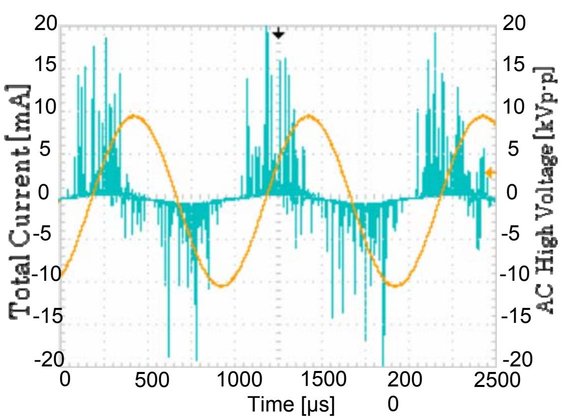


Figure 6 Waveform of AC high voltage and Total current for generation of the uniform slide discharge
(Negative DC high voltage -12.5kV, AC high voltage 20kVp-p, 2.0 mm inner diameter, 20 mm capillary)

Figure 7 and Figure 8 show the waveforms of the AC high voltage and the total current with higher AC voltage and lower DC voltage. The maximum potential differences were nearly the same in all the experiments (22.5kV-23kV). The AC and DC voltages were 25kVp-p and -10kV in figure 8 and 30kVp-p and -8kV in figure 8. Figures 6, 7 and 8 show that amplitude of the current pulse associated with the honeycomb discharge is nearly the same in these figures. The current pulses associated with the honeycomb discharge started at a different value of AC voltages but at nearly the same AC+DC voltages (20kV). These results suggest that honeycomb discharge is initiated when the electric field exceeds a certain value. The threshold value was about 10kV/cm in these experimental conditions.

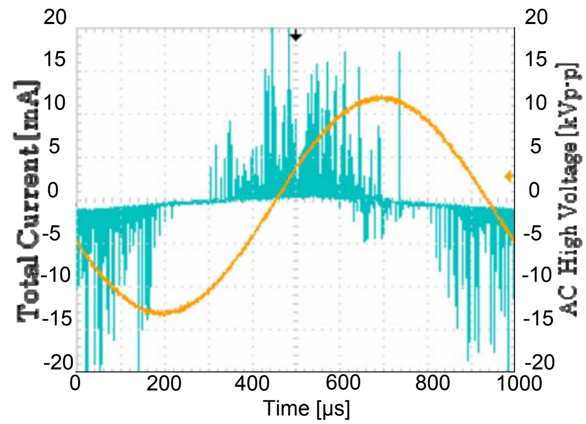


Figure 7 Waveform of AC high voltage and Total current for generation of the uniform slide discharge
(Negative DC high voltage -10kV, AC high voltage 25kVp-p, 2.0 mm inner diameter, 20 mm capillary)

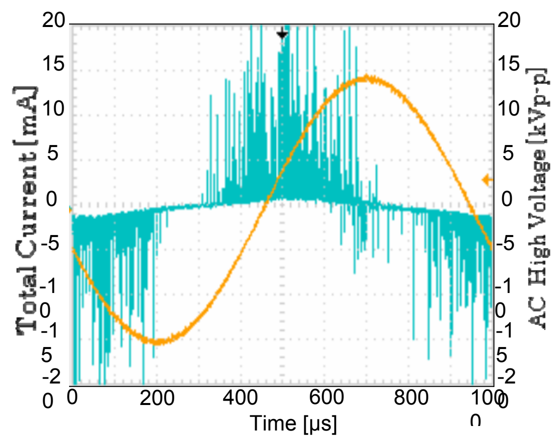


Figure 8 Waveform of AC high voltage and Total current for generation of the uniform slide discharge
(Negative DC high voltage -8kV, AC high voltage 30kVp-p, 2.0 mm inner diameter, 20 mm capillary)

D. Emission spectra of the Honeycomb Discharge in bundle of capillaries

Figure 9 indicates the DC high voltage vs. emission of the honeycomb discharge. The emission of honeycomb discharge was measured using a spectrometer (Ocean Optics SD2000 and USB4000). The peak value of the measured emission of 337.01nm (N_2 C-B,0-0) is plotted.

The inner diameter was 2mm and the length was 20mm of the bundled of quartz glass capillaries. AC frequency was set to 50Hz. The applied AC high voltage was set 20kV_{p-p}, 25kV_{p-p}, 30kV_{p-p}, 50Hz, and the DC high voltage was varied when measuring the emission of honeycomb discharge.

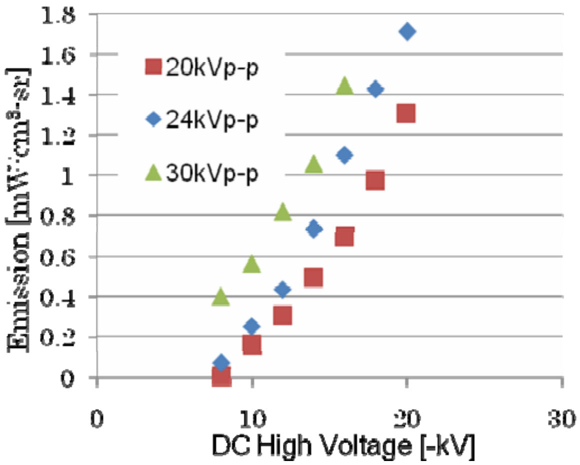


Figure 9 DC high voltages vs. Emission
(AC50Hz, 2.0 mm inner diameter, 20 mm capillary)

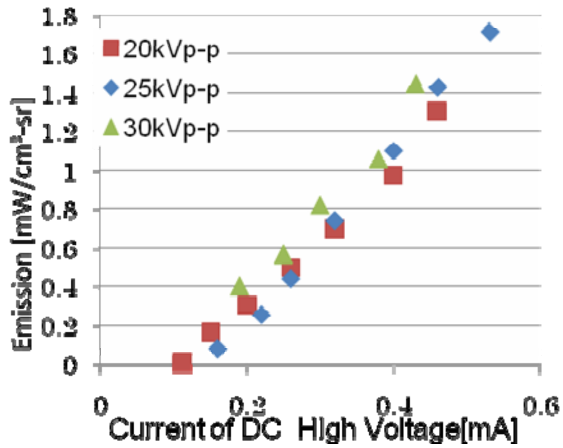
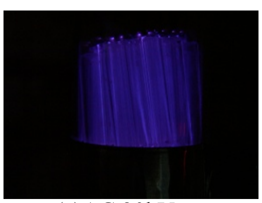


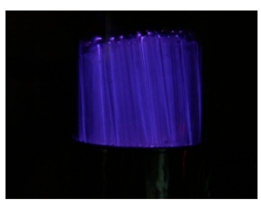
Figure 11 DC Current vs. Emission
(AC50Hz, 2.0 mm inner diameter, 20 mm capillary)

With the same ac voltage, the emission was stronger with increasing dc voltages. At the same DC voltage, the emission was stronger with increasing AC voltages.

Figure 10 shows photographs of the honeycomb discharge when the light emission was measured. The DC voltage was -16 kV. AC voltage of each photograph is as follows: (a): 20kVp-p, (b): 25kVp-p, (c): 30kVp-p, to form a honeycomb discharge in the bundle. Using the transparent quartz glass tube, generation of the ionization inside these tubes was confirmed.



(a)AC 20kV$_{p\text{-}p}$



(b)AC 25kV$_{p\text{-}p}$

(c)AC 30kV$_{p\text{-}p}$

Figure 10 Photographs of honeycomb discharge
(DC high voltage -16kV)

In case of each AC high voltage, quartz glass capillaries were confirmed that an electric discharge with the emission of light occurs uniformly. In addition, light emission was confirmed that was stronger along AC high voltage value.

Figure 11 shows the light emission intensity vs. DC current. The AC high voltage was set to 20kV$_{p\text{-}p}$, 25kV$_{p\text{-}p}$, 30kV$_{p\text{-}p}$, 50Hz, and the DC high voltage was changed when measuring the emission. The light emission seems proportional to the DC current.

Unlike the relations between the DC high voltage and emission, the DC high voltage current was have risen with the value of DC high voltage regardless of a value of the AC high voltage. The emission spectrum of the honeycomb discharge depended on the current of the DC high voltage was suggested.

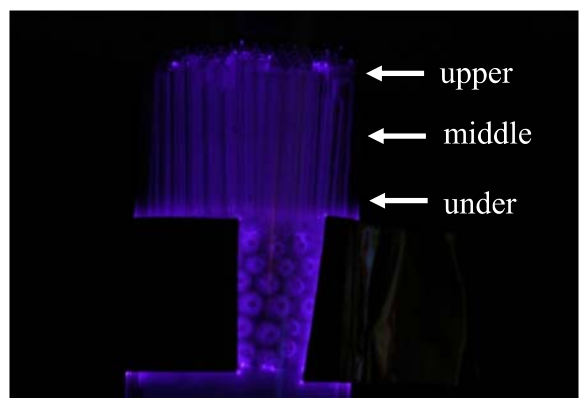


Figure 12 Photographs of honeycomb discharge
(AC 25kV$_{p\text{-}p}$ 1kHz, DC -15kV 2mA, N_2 gas 3L/min,
2.0 mm inner diameter, 20 mm capillary)

Figure 12 shows photographs of the honeycomb discharge when the light emission was measured. The AC high voltage was set to 25kV$_{p\text{-}p}$, 1kHz and the DC high voltage was set to -15kV. The gas flow was set to 3L/min N_2 gas.

The emission Intensity was measured at the upper, middle and under side as indicated in Figure 12m and Figure 13 shows the wave length vs. light emission intensity at those parts. The AC high voltage was set to 25kV$_{p\text{-}p}$, 1kHz and the DC high voltage was set to -15kV. The gas flow was set to 3L/min N_2 gas.

The emission intensity of the under-side was the strongest. Next was the middle (89% of the under-side) and the lowest was from the upper-side(81%). This result showed the attenuation of the light emission was 10% at 1cm.

These results suggested that the emission spectrum and intensity of the honeycomb discharge depended on the length of honeycomb.

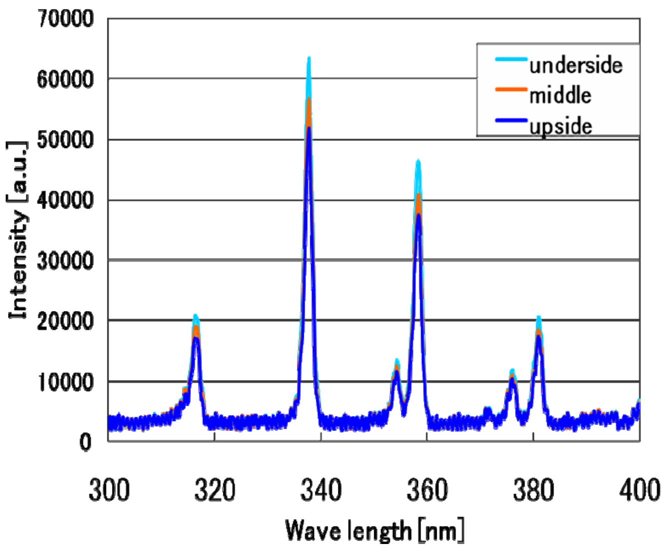


Figure 13 Wave length vs. Intensity
(AC 25kV$_{p-p}$ 1kHz, DC -15kV 2mA, N_2 gas 3L/min, 2.0 mm inner diameter, 20 mm capillary)

IV. Conclusion

Using both AC and DC high voltage, generation of plasma inside a honeycomb has become possible. Using a bundle of transparent capillaries simulating honeycomb catalyst, characteristics of the honeycomb discharge has been studied.

AC voltage was applied to the packed bed, and DC voltage was applied to the bundle. To generate a stable honeycomb discharge, the condition of the AC and DC voltages are clarified as the onset and the flashover voltages. The average electric field strength for the onset of the honeycomb discharge was about 10kV/cm in this experimental condition. The honeycomb discharge is pulsating, detected as the current pulses at an instant of the maximum of the AC voltage. The light emission intensity from the honeycomb discharge is roughly in proportion to the DC current.

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